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BF_3·OEt_2-Promoted Annulation for Substituted 2-Arylpyridines as Potent UV Filters and Antibacterial Agents

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Abstract. A simple and efficient BF_3·OEt_2 mediated methodology for the construction of diverse 2-phenylpyridines bearing benzophenone moieties from readily available 3-formylchromones and phenylacetylenes in wet acetonitrile was developed. The nitrogen source for the pyridine construction was derived from acetonitrile. This one-pot protocol proceeds *via* [3+2+1] annulation through cascade nucleophilic addition, hydrolysis, Michael-type addition, ring opening, and elimination reactions. The synthesized compounds may have applications as UV filters and exhibit potent antibacterial activities.

Keywords: BF_3·OEt_2; 2-phenylpyridines; [3+2+1] annulation; UV filters; Antibacterial

Introduction

2-Arylpyridines are key skeletons in pharmaceuticals and bioactive materials (Figure 1).[1] In addition, they are also used as starting materials or building blocks in the synthesis of valuable ligands for metal complexes[2] and in important organic transformations.[3] Owing to the importance of 2-arylpyridines, several synthetic approaches have been reported based on transition-metal catalyzed or metal-free conditions. Typical methods for the synthesis of 2-arylpyridines include copper-catalyzed C-N bond activation of acetophenones with 1,3-diaminopropane,[4] nucleophilic addition of a dithiane anion to an *α,β*-unsaturated carbonyl followed by the metallacycle-mediated union of allylic alcohols with pre-formed trimethylsilane-imines,[5] copper-catalyzed coupling of oxime carboxylates to vinylboronic acids,[6] condensation of enaminones with ammonium acetate,[7] cyclocondensation of 3-chloro-1-phenyl-pro-2-enone with 3-amino-1-phenyl pro-2-enone,[8] and reaction of ethyl acetoacetate with phenyl-2-propyn-1-one and ammonium acetate.[9] Importantly, the nitrogen source for 2-arylpyridine formation was derived from 1,3-diaminopropane lithium hexamethyldisilazide, oxime carboxylate, ammonium acetate, and ammonia.

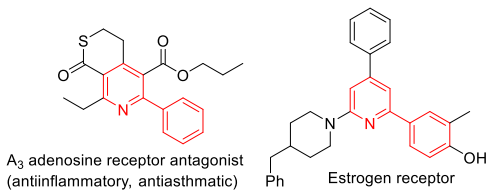
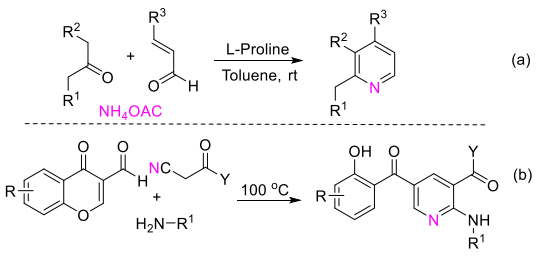


Figure 1. Selected bioactive molecules bearing 2-arylpyridine moieties.

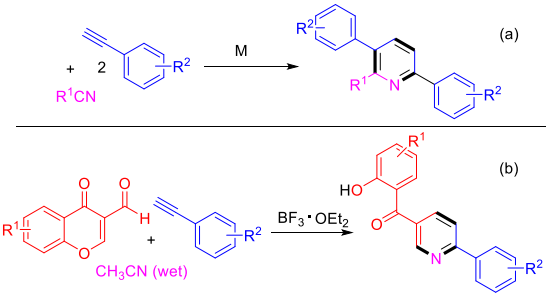
Although a number of approaches for the synthesis of 2-arylpyridines have been developed,[10] a more facile and efficient one-pot synthetic protocol is still desirable. Recently, we reported an organocatalyzed oxidative *N*-annulation for the synthesis of pyridines from ketones with *α,β*-unsaturated aldehydes and ammonium acetate (method a, Scheme 1).[11] We also described a catalyst- and solvent-free thermal multicomponent cascade of 4-oxo-4*H*-chromene-3-carbaldehydes with cyanoacetates and anilines for the



Scheme 1. Our reported synthetic strategies for pyridines.

synthesis of 2-aminopyridines (method b, Scheme 2).[12]

Recently, metal-mediated or catalyzed [2+2+2] cycloaddition of two alkynes with nitriles has become one of the most powerful strategies for the construction of substituted pyridines (method a, Scheme 2).[13] In these cases, nitriles were used as an active reagent for the formation of pyridine ring. To date, there are no reports on [3+2+1] annulations for the synthesis of 2-arylpyridines using acetonitrile as a nitrogen source. As part of our ongoing interest in this area, we report a novel $BF_3{\cdot}OEt_2$ mediated [3+2+1] annulation of commercially available 3-formylchromones, aryl acetylenes, and acetonitrile for the construction of diverse 2-arylpyridines (method b, Scheme 2). Interestingly, in this reaction, the nitrogen source for the 2-arylpyridine construction is from acetonitrile, rather than ammonium acetate or the amine.

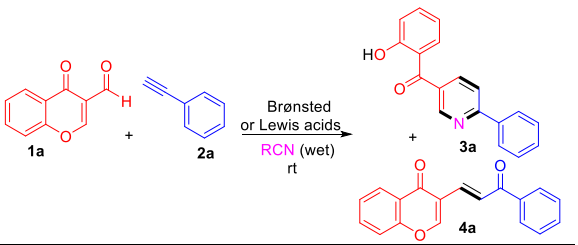


Scheme 2. Strategy for the construction of 2-arylpyridines.

Results and Discussion

The reaction of 3-formylchromone (**1a**) with phenylacetylene (**2a**) in wet nitriles was first investigated using several Brønsted and Lewis acids (Table 1). Initial attempts with Brønsted acids such as *p*-TsOH and TFA in wet acetonitrile at room temperature for 12 h did not furnish the desired product (entries 1 and 2). Several Lewis acids such as AgOTf, $Cu(OTf)_2$, $In(OTf)_3$, $InCl_3$, $Fe(OTf)_3$, $Y(OTf)_3$, $TiCl_4$, $SnCl_4$, $AlCl_3$, $FeCl_3$ and $BF_3{\cdot}OEt_2$ were next screened (entries 3-13). Among these, $BF_3{\cdot}OEt_2$ (1.0 equiv.) gave **3a** in 58% isolated yield (entry 13). With other Lewis acids, only starting material **1a** was recovered. Decreasing the amount of $BF_3{\cdot}OEt_2$ to 0.5 equivalent did not improve the yield (entry 14). When the amount of $BF_3{\cdot}OEt_2$ was increased to 2.0 equivalent, product **3a** was obtained in 79% yield (entry 15). However, when the reaction was screened with other nitriles, two products **3a** and **4a** were isolated. For example, treatment of **1a** with **2a** in wet propionitrile provided **3a** in 28% yield with trace amount of **4a** (entry 16). With other nitriles such as valeronitrile, methoxypropionitrile, benzonitrile, and phenylacetonitrile, both **3a** (31-41%) and **4a** (trace-18%) were produced (entries 17-20). In other solvents having no nitrogen source such as 1,2-dichloroethane and ethanol, no products were produced and starting materials were recovered (entries 21-22). Importantly, the reaction was also carried out in dry acetonitrile, desired product **3a** was not obtained, instead, intractable mixture was produced (Scheme 4). The structures of **3a** and **4a** were confirmed by spectroscopic analyses. The ^1H NMR spectrum of **3a** showed a characteristic -OH peak at δ = 11.89 ppm as a singlet and one aromatic peak on the pyridine moiety at δ = 8.97 ppm (J = 2.4 Hz) as a doublet due to long range coupling. Compound **4a** showed a characteristic vinylic proton peak on the chromenone ring at 8.19 ppm as a singlet and two *trans*-vinylic protons at δ = 8.66 ppm (J = 15.0 Hz) and 7.47 ppm (J = 15.0 Hz) as two doublets. The structure of **3a** was further confirmed by X-ray crystallographic analysis of related compound **3e**.

Table 1. Optimization of reaction conditions for the synthesis of **3a**.[a]



Entry	Brønsted or Lewis acids	Solvents	Time (h)	Yield (%)[b] 3a	Yield (%)[b] 4a
1	*p*-TsOH (1.0 eq)	CH_3CN	12	0	0
2	TFA (1.0 eq)	CH_3CN	12	0	0
3	AgOTf (1.0 eq)	CH_3CN	12	0	0
4	$Cu(OTf)_2$ (1.0 eq)	CH_3CN	12	0	0
5	$In(OTf)_3$ (1.0 eq)	CH_3CN	12	0	0
6	$InCl_3$ (1.0 eq)	CH_3CN	12	0	0
7	$Fe(OTf)_3$ (1.0 eq)	CH_3CN	12	0	0
8	$Y(OTf)_3$ (1.0 eq)	CH_3CN	12	0	0
9	$TiCl_4$ (1.0 eq)	CH_3CN	12	0	0
10	$SnCl_4$ (1.0 eq)	CH_3CN	12	0	0
11	$AlCl_3$ (1.0 eq)	CH_3CN	12	0	0
12	$FeCl_3$ (1.0 eq)	CH_3CN	12	0	0
13	$BF_3{\cdot}OEt_2$ (1.0 eq)	CH_3CN	1	58	0
14	$BF_3{\cdot}OEt_2$ (0.5 eq)	CH_3CN	1	32	0
15	$BF_3{\cdot}OEt_2$ (2.0 eq)	CH_3CN	1	79	0
16	$BF_3{\cdot}OEt_2$ (2.0 eq)	CH_3CH_2CN	1	28	trace
17	$BF_3{\cdot}OEt_2$ (2.0 eq)	$CH_3(CH_2)_3CN$	1	31	18
18	$BF_3{\cdot}OEt_2$ (2.0 eq)	$CH_3OCH_2CH_2CN$	1	39	trace
19	$BF_3{\cdot}OEt_2$ (2.0 eq)	PhCN	1	41	13
20	$BF_3{\cdot}OEt_2$ (2.0 eq)	$PhCH_2CN$	1	39	18
21[c]	$BF_3{\cdot}OEt_2$ (2.0 eq)	1,2-Dichloroethane	1	0	0
22[d]	$BF_3{\cdot}OEt_2$ (2.0 eq)	CH_3CH_2OH	1	0	0

[a]Reaction conditions: **1a** (0.6 mmol), **2a** (0.66 mmol), and solvent (1 mL). [b]Isolated yield.*[c],[d] = without nitrogen source

Next, the generality of the reaction was explored using diverse 3-formylchromones **1b-1h** with phenylacetylene (**2a**) under the optimized conditions (Table 2). For example, treatment of **1b-1d** bearing electron-donating groups (6-Me, 6-Et, and 6-*i*Pr) with **2a** in wet acetonitrile at room temperature for 1 h provided the desired products **3b-3d** in 79%, 76%, and 75% yields, respectively, whereas **1e-1g** bearing electron-withdrawing groups (6-Br, 6-Cl, and 6-F) afforded **3e-3g** in 72%, 72%, and 68% yields, respectively. Notably, 3-formylchromone **1h** bearing both electron-donating and electron-withdrawing groups on the aromatic ring were also transformed into desired product **3h** in 70% yield.

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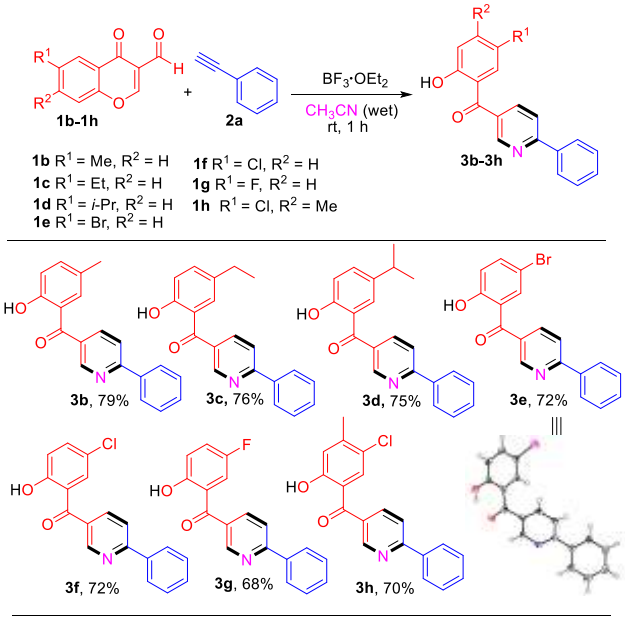
Table 2. Substrate scope of 3-formylchromones **1b-1h** in formation of 2-phenylpyridines **3b-3h**.



To demonstrate the versatility of the developed reaction, various arylacetylenes were tested (Table 3). The reactions proceeded smoothly to provide the corresponding products in good yields. For example, when 3-formylchromone (**1a**) was treated with arylacetylenes **2b-2e** bearing electron-donating groups such as 3-methyl, 4-methyl, 4-*n*-propyl, and 4-*n*-pentyl in acetonitrile, the desired products **5a-5d** were obtained in 70-78% yields. Similarly, with arylacetylenes **2f-2h** bearing electron-withdrawing groups 4-Br, 4-CF_3, and 3,5-CF_3 on the benzene ring, **5e**, **5f**, and **5g** were isolated in 73%, 69%, and 68% yields, respectively. The reaction of **1a** with 1,4-diethynylbenzene (**2i**) in wet acetonitrile gave **5h** in 71% yield without concomitant formation of the bispyridine. With 1-ethynylnaphthalene (**2j**), **5i** bearing a naphthyl ring was formed in 70% yield.

The scope of the substrates was further extended by employing substituted 3-formylchromones (**1b-1c** and **1e-1i**) and substituted phenylacetylenes (**2c-2i** and **2k-2l**) (Table 4). Treatment of 6-methyl-3-formylchromone (**1b**) with 1-ethynyl-4-methoxy-2-methylbenzene (**2k**) or 1-ethynyl-3,5-bis(trifluoromethyl)benzene (**2h**) in wet acetonitrile at room temperature for 1 h gave **6a** and **6b** in 76% and 68% yields, respectively; the reaction of **1c** with **2c** provided **6c** in 75% yield. Similarly, halogen-substituted 3-formylchromones **1e-1g** with substituted phenylacetylenes **2c-2g** and **2l** provided the corresponding products **6d-6i** in 69-75% yields. In addition, the combination of dichloride-substituted 3-formylchromones **1i** with **2c** or **2i** afforded products **6j** and **6k** in 74% and 71% yields, respectively. 3-Formylchromone **1h** with **2g** was also transformed into **6l** in 70% yield. This methodology provides a rapid route towards diverse 2-arylpyridines bearing benzophenones.

Table 3. Substrate scope of phenylacetylenes **2b-2j** in formation of 2-arylpyridines **5a-5i.**

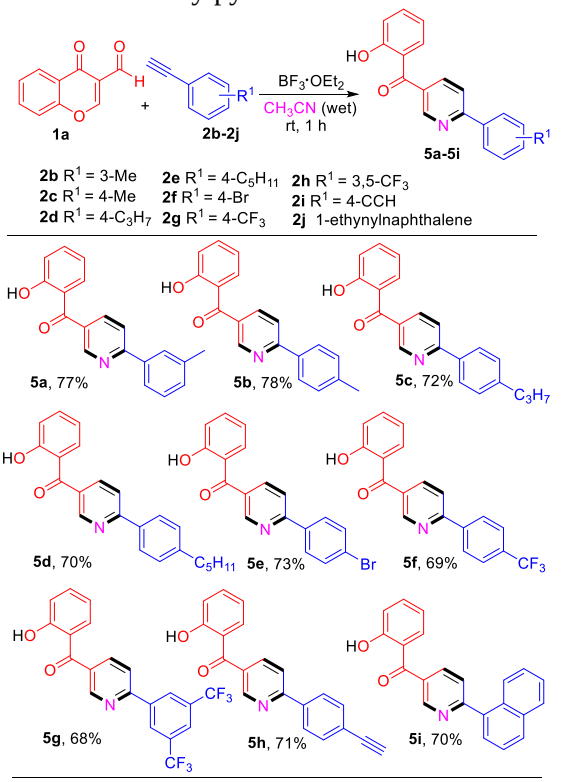
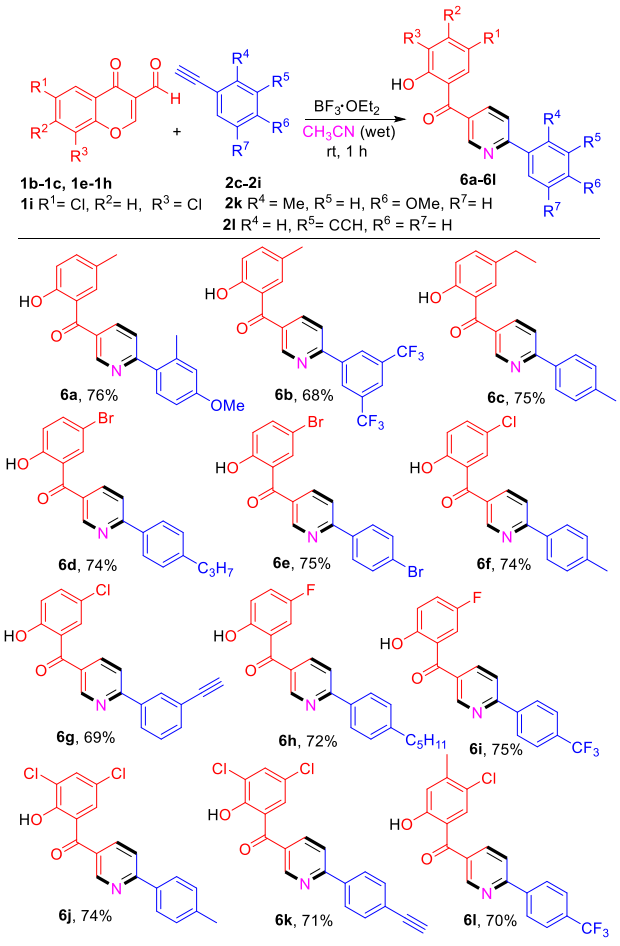
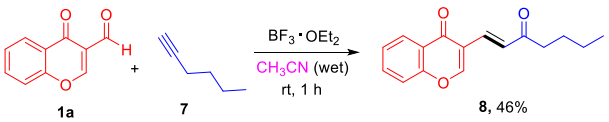


Table 4. Substrate scope of 3-formylchromones **1b-1c**, **1e-1i** and arylacetylenes **2c-2i** and **2k-2l** in formation of 2-arylpyridines **6a-6l.**

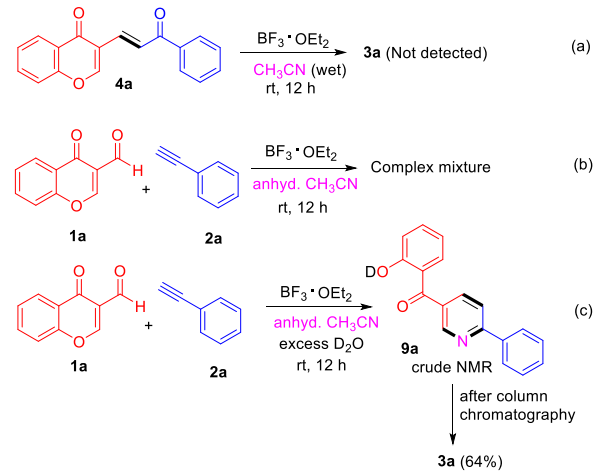


Having seen the generality and versatility of the reaction, the possibility of use of aliphatic alkyne was next examined. For example, the reaction of **1a** with 1-hexyne (**7**) in the presence of 2 equivalents of $BF_3{\cdot}OEt_2$ in wet acetonitrile did not provide the desired product **3a**, instead compound **8** was isolated in 46% yield (Scheme 3).



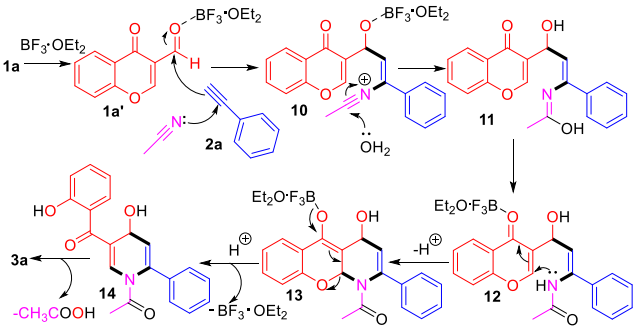
Scheme 3. Reaction of **1a** with 1-hexyne (**7**).

To gain insight into the mechanism, some control experiments were carried out (Scheme 4). The reaction of compound **4a** was carried out under standard conditions, desired cycloadduct **3a** was not formed and only starting material **4a** was recovered (eq. a, Scheme 4). When the reaction was performed under argon atmosphere with anhydrous acetonitrile, intractable mixture was obtained (eq. b, Scheme 4). Further reaction between **1a** with **2a** in anhydrous acetonitrile with excessive D_2O was performed, deuterium exchanged compound **9a** was obtained, which is detected in crude ^1H NMR (method c, Scheme 4). However, after purification by column chromatography, only compound **3a** was isolated in 64% yield. These observations showed that compound **4a** is a side product instead of intermediate and that water is preferentially necessary to produce the desired product **3a**.



Scheme 4. Control Experiments.

On the basis of above mentioned control experiments and results,[14] a possible mechanism for the formation of **3a** is depicted in Scheme 5. In the presence of $BF_3{\cdot}OEt_2$, 3-formylchromone (**1a)** gives complex **1a'**. The addition of acetonitrile to **2a** followed by nucleophilic addition to **1a'** affords intermediate **10**. Hydrolysis of nitrile group followed by keto-enol tautomerization of **11** generates intermediate **12**. Intramolecular Michael-type addition of the nitrogen on amide moiety gives **13** which undergoes retro Michael-type ring opening furnishes intermediate **14.** Elimination of acetic acid from **14** finally affords product **3a**.



Scheme 5. Proposed mechanism for the formation of **3a**.

The optical and chemical properties of the compounds bearing benzophenone moieties have important implications in photochemistry.[15] As such, the potential of the synthesized 2-arylpyridines bearing benzophenone skeletons as UV filters was explored *in vitro*. Although benzophenones have been widely used as potent ingredients in sunscreen, the development of new UVA/UVB filters is required owing to the prevalence of allergic reactions and free radical damage by benzophenone derivatives.[15b,16] Therefore, the UV absorption of **3a**, **3b**, **3f**, **3h**, **5a**, **5e**, **5f**, **6b**, **6g**, and **6j** were evaluated to determine their viability and performance as UV filters (Figure 2). Specifically, the molar extinction coefficient (ε), UVA/UVB ratio, and critical wavelength were determined.

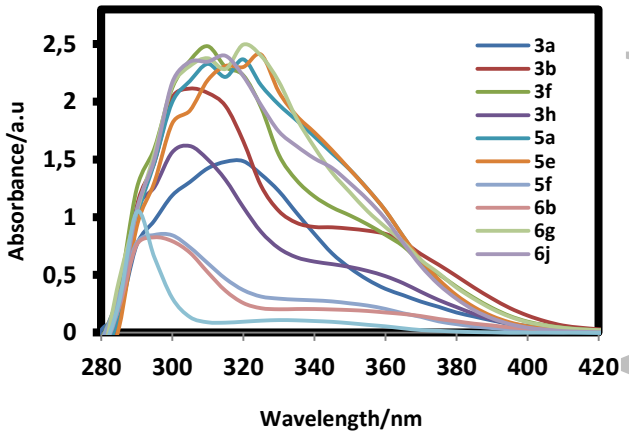


Figure 2. Absorbance *vs.* wavelength of **3a**, **3b**, **3f**, **3h**, **5a**, **5e**, **5f**, **6b**, **6g**, and **6j** in EtOH.

The molar extinction coefficient (ε), which is an important variable that describes the light absorbance strength of a substance at a given wavelength per molar concentration, was computed using Beer-Lambert's law. The UVA/UVB ratio and critical

wavelength (λ_c) were determined using the method described by Diffey (Table 5).[17] According to previous studies, the prerequisite for broad-spectrum UVA/UVB protection labeling is satisfied when the critical wavelength is greater than 370 nm.[18]

Table 5. UV absorption properties of synthesized compounds.

Compounds[a]	Λ_{nm}	UVA/UVB	Λ_c	ε (L.mol^{-1}cm^{-1})
3a	320	0.43	359	18355
3b	305 340	0.41	371	24438 10577
3f	310 320	0.42	365	30665 27550
3h	304 340	0.33	364	20736 7946
5a	310 320	0.53	362	26912 27363
5e	310 320	0.53	362	32964 33517
5f	295 340	0.27	360	11607 3828
6b	295 340	0.25	366	16104 3982
6g	310 320	0.51	367	31662 33233
6j	315 340	0.46	361	26378 16599
Oxybenzone	288 326	0.17	349	6665 949

[a]The concentration of tested sample was 50 μM in EtOH.

Although **3a**, **3f**, **3h**, **5a**, **5e**, **5f**, **6b**, **6g**, and **6j** showed a broad spectrum in the UVB region, only **3b** exhibited a critical wavelength (371 nm) to be classified as a broad-spectrum protection agent. In terms of the UVA/UVB ratio, which is based on the mean absorbance in the UVA region divided by the mean absorbance in the UVB region,[19] compounds **5a**, **5e**, and **5g** showed the highest values of 0.53, 0.53, and 0.51, respectively, indicating good UVA-coverage compared to UVB-coverage. Shifts in the absorbance towards the blue or red region could have been caused by the substituents present on the phenyl group attached to the pyridine ring of the compounds. For instance, compounds **3b**, **3h**, **5f**, and **6b** showed hypsochromic (blue) shifts, while compounds **5e** and **6g** exhibited bathochromic (red) shifts in their λ_{max}, which were all based on **3a** (Figure 2).

Since pyridine moieties have central roles in many biological activities, the antibacterial activities of some selected synthetic compounds (**3h**, **5f**, **5g**, **6b**, and **6e**) were screened against gram-negative bacteria *E. coli* (KCTC-1924) and *P. aeruginosa* (KCTC-2004), as well as gram-positive bacteria *S. aureus* (KCTC-1916) and *B. cereus* (KCTC-1012) using a modified version of the Kirby-Bauer disk diffusion method.[20] The bacterial strains were obtained from the Korean Collection for Type Cultures (KCTC). The bacterial strains were cultivated in 10 mL of fresh Difco™ broth for 24 h. The optical density of the newly cultivated bacterial suspension was 0.7 at 595 nm. 100 μL aliquots from the bacterial suspension were then spread over the Difco™ nutrient broth (prepared *via* the method described above). Filter paper disks with 8 mm diameters, previously autoclaved and saturated with 20 μL of the test sample, were placed on the agar plate and incubated for 20-36 h at 37 °C. After the incubation period, the zones of inhibition were measured. Standard discs of ciprofloxacin were used as positive controls. Filter paper discs impregnated with DMSO were used as negative controls. Notably, the depth of the agar in the plate must be considered in the disc diffusion method.

The antibacterial activities of the compounds are recorded in Table 6. When filter paper discs impregnated with the chemical compounds were placed on the agar medium, the chemical compounds diffused into the agar. The solubility of the chemical compound and its molecular mass determines the infiltration area. When bacteria are placed on the agar plate, they will not grow in the area around the disc if the chemical compound present on the disc has antibacterial activity. The area (diameter) of no growth around the disc is known as the zone of inhibition. A few of the compounds *viz*. **3h**, **5f**, **5g**, **6b**, and **6e** were selectively active against gram positive and gram negative bacteria. Compounds **3h**, **5f**, **5g**, **6b**, and **6e** appeared to be moderately active against most of the strains as compared to the reference antibiotic (ciprofloxacin). Compound **5g** exhibited a zone of inhibition with a diameter of 12 mm against *P. aeruginosa*, 15 mm against *E. coli*, 10 mm against *S. aureus*, and 15 mm for *B. cereus*. Furthermore, compound **6b** cast zones of inhibition with diameters of 10, 12, 11, and 14 mm for *P. aeruginosa*, *E. coli*, *S. aureus*, and *B. cereus*, respectively, indicating that the synthesized compounds bearing strong electron withdrawing groups on the phenyl ring showed better antibacterial activities than other compounds.

Table 6. Antibacterial activity of tested compounds against several standard bacterial strains.

Entry	Compound	Diameter of growth inhibition zone (mm)			
		Gram-negative		Gram-positive	
		P. aeruginosa	*E.Coli*	*S. aureus*	*B. cereus*
1	**3h**	10	15		12
2	**5f**		14	10	10
3	**5g**	12	15	10	15
4	**6b**	10	12	11	14
5	**6e**		10		12
Standard ciprofloxacin		18	19	18	20

Conclusion

In conclusion, we have developed a facile and efficient methodology for the construction of

substituted 2-arylpyridines bearing benzophenone moieties starting from commercially available 3-formylchromones and arylacetylenes in acetonitrile with good yields. This protocol offers mild reaction conditions and a convenient one-pot procedure. Moreover, many functional groups on the benzene rings are tolerated. The synthesized compounds were found to have applications as UV protecting materials, especially compound **3b**, which exhibited the most potential with a critical wavelength of 371 nm. As for the antibacterial applications, the tested compounds showed moderate activities, and further improvements may be possible.

Experimental Section

General remarks:

All experiments were carried out in open air. Merck precoated silica gel plates (Art. 5554) with a fluorescent indicator were used for analytical TLC. Flash column chromatography was performed using silica gel 9385 (Merck). The melting points are uncorrected and were determined using micro-cover glasses on a Fisher-Johns apparatus. ^{1}H NMR spectra were recorded on a Varian-VNS (600 MHz) spectrometer using the chemical shift of the solvent at δ = 7.24 ppm for $CDCl_3$ or δ = 0.00 ppm for TMS as a reference. ^{13}C NMR spectra were recorded on a Varian-VNS (150 MHz) spectrometer using the chemical shift of the solvent at δ = 77.0 ppm for $CDCl_3$ as a reference. Chemical shifts (δ) are expressed in units of ppm and J values are given in Hz. Multiplicities are abbreviated as follows: s = singlet, d = doublet, t = triplet, q = quartet, br s = broad singlet, dd = doublet of doublets, td = triplet of doublets, quint = quintet, sept = septet, and m = multiplet. IR spectra were recorded on a FTIR (BIO-RAD) and high-resolution mass spectra were obtained on a JEOL JMS-700 spectrometer at the Korea Basic Science Institute.

General procedure:

To a mixture of 3-formylchromone (**1**) (0.6 mmol) and phenylacetylene (**2**) (0.66 mmol, 1.1 equiv.) in acetonitrile (1 mL) was added boron trifluoride etherate (0.6 mmol, 1 equiv.) at room temperature; the mixture was stirred for 1 h. The progress of the reaction was monitored by TLC. After the completion of the reaction, the solvent was removed on a rotary evaporator and the product was extracted with ethyl acetate (2 × 10 mL) and then washed with brine (5 mL). The organic layer was dried over Na_2SO_4 and evaporated to give the crude product, which was purified by short column chromatography over silica gel (EtOAc/hexane = 1:9) to give the desired products.

Characterization data of synthesized compounds:

(2-Hydroxyphenyl)(6-phenylpyridin-3-yl)methanone (3a). The title compound (**3a**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 119-121 °C. Yield: 79% (130 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.90 (1H, s), 8.97 (1H, d, J = 2.4 Hz), 8.08-8.05 (3H, m), 7.86 (1H, d, J = 7.8 Hz), 7.61 (1H, d, J = 8.4 Hz), 7.54-7.45 (4H, m), 7.08 (1H, d, J = 8.4 Hz), 6.90 (1H, t, J = 7.8 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 199.0, 163.2, 160.0, 149.9, 137.9, 137.6, 136.8, 132.9, 131.6, 130.0, 128.9, 127.3, 119.8, 119.0, 118.9, 118.6; IR (ATR) 3023, 1737, 1619, 1583, 1477, 1445, 1385, 1304, 1242, 1209, 1140, 1016, 786 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{18}H_{13}NO_2$: 275.0946. Found: 275.0948.

(2-Hydroxy-5-methylphenyl)(6-phenylpyridin-3-yl) methanone (3b). The title compound (**3b**) was prepared according to the general procedure. The product was obtained as a brown solid, mp 89-90 °C. Yield: 78% (135 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.71 (1H, s), 8.97 (1H, d, J = 4.4 Hz), 8.08-8.06 (3H, m), 7.87 (1H, d, J = 3.9 Hz), 7.51-7.45 (3H, m), 7.38 (1H, s), 7.34 (1H, dd, J = 9.0, 2.4 Hz), 6.99 (1H, d, J = 8.4 Hz), 2.25 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.9, 161.1, 159.9, 149.9, 137.9, 137.8, 137.7, 132.5, 131.9, 130.0, 128.9, 128.2, 127.3, 119.9, 118.7, 118.4, 20.4; IR (ATR) 2919, 1626, 1585, 1475, 1384, 1294, 1224, 1141, 1020, 858, 770 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{19}H_{15}NO_2$: 289.1103. Found: 289.1102.

(5-Ethyl-2-hydroxyphenyl)(6-phenylpyridin-3-yl)methanone (3c). The title compound (**3c**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 115-117 °C. Yield: 76% (138 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.71 (1H, s), 8.98 (1H, d, J = 4.4 Hz), 8.10-8.07 (3H, m), 7.89 (1H, d, J = 3.9 Hz), 7.52-7.46 (3H, m), 7.40-7.38 (2H, m), 7.02 (1H, d, J = 8.4 Hz), 2.56 (2H, q, J = 7.8 Hz), 1.17 (3H, t, J = 7.8 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.9, 161.3, 159.9, 149.9, 137.9, 137.8, 136.9, 134.8, 131.9, 131.5, 130.1, 128.9, 127.4, 120.1, 118.8, 118.5, 27.9, 15.8; IR (ATR) 2964, 2918, 2858, 1585, 1478, 1411, 1386, 1338, 1133, 1109, 778 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{20}H_{17}NO_2$: 303.1259. Found: 303.1256.

(2-Hydroxy-5-isopropylphenyl)(6-phenylpyridin-3-yl)methanone (3d). The title compound (**3d**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 155-157 °C. Yield: 75% (142 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.72 (1H, s), 8.99 (1H, d, J = 1.2 Hz), 8.10-8.08 (3H, m), 7.89 (1H, d, J = 7.8 Hz), 7.53-7.46 (3H, m), 7.44-7.42 (2H, m), 7.03 (1H, d, J = 8.4 Hz), 2.83 (1H, sept, J = 7.2 Hz), 1.19 (6H, d, J = 7.2 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.9, 161.4, 160.1, 149.9, 139.5, 138.0, 137.8, 135.4, 131.9, 130.2, 130.1, 128.9, 127.4, 120.1, 118.7, 118.5, 33.3, 23.9; IR (ATR) 2917, 2849, 1622, 1575, 1473, 1329, 1298, 1236, 1145, 1105, 1068, 1004, 813, 753 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{19}NO_2$: 317.1416; Found: 317.1412.

(5-Bromo-2-hydroxyphenyl)(6-phenylpyridin-3-yl)methanone (3e). The title compound (**3e**) was prepared according to the general procedure. The product was obtained as a yellow solid and crystallized in ethanol, mp 157-159 °C. Yield: 72% (152 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.77 (1H, s), 8.98 (1H, d, J = 1.8 Hz), 8.09 (3H, dd, J = 8.4, 1.8 Hz), 7.91 (1H, d, J = 8.4 Hz), 7.72 (1H, d, J = 2.4 Hz), 7.61 (1H, dd, J = 9.0, 2.4 Hz), 7.53-7.47 (3H, m), 7.00 (1H, d, J = 9.0 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 197.9, 162.1, 160.5, 149.9, 139.5, 137.8, 137.7, 134.7, 131.1, 130.3, 129.0, 127.4, 120.7, 120.3, 120.1, 110.7; IR (ATR) 3062, 1626, 1586, 1460, 1381, 1319, 1289, 1154, 1081, 1020, 939, 831, 691 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{18}H_{12}BrNO_2$: 353.0051. Found: 353.0048.

X-Ray crystallographic data of compound 3e: Empirical Formula- $C_{23}H_{22}N_2O_4$, M = 354.20, Monoclinic, Space group $P2_1/c$, a = 6.2873(3) Å, b = 16.9635(8) Å, c = 13.8579(7) Å, V = 1448.39(12) Å^3, Z = 4, T = 223(2) K, ρ_{calcd} = 1.624 Mg/m^3, $2\Theta_{max.}$ = 28.357°, Refinement of 200 parameters on 3622 independent reflections out of 46289 collected reflections (R_{int} = 0.0422) led to R_1 = 0.0299 [$I > 2\sigma(I)$], wR_2 = 0.0738 (all data) and S = 1.076 with the largest difference peak and hole of 0.584 and -0.615 e. Å^{-3} respectively. The crystal structure has been deposited at the Cambridge Crystallographic Data Centre (CCDC 1570166). The data can be obtained free of charge via the Internet at www.ccdc.cam.ac.uk/data_request/cif.

(5-Chloro-2-hydroxyphenyl)(6-phenylpyridin-3-yl)methanone (3f). The title compound (**3f**) was prepared according to the general procedure. The product was obtained as a brown solid, mp 171-173 °C. Yield: 70%

(129 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.75 (1H, s), 8.98 (1H, d, J = 2.4 Hz), 8.09 (3H, dd, J = 8.4, 1.8 Hz), 7.90 (1H, d, J = 8.4 Hz), 7.58 (1H, d, J = 3.0 Hz), 7.53-7.47 (4H, m), 7.05 (1H, d, J = 9.6 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.0, 161.6, 160.5, 149.9, 137.8, 137.7, 136.7, 131.7, 131.1, 130.3, 129.0, 127.4, 123.8, 120.4, 120.1, 119.7; IR (ATR) 3074, 1627, 1587, 1461, 1384, 1320, 1208, 946, 834, 694 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{18}H_{12}ClNO_2$: 309.0557. Found: 309.0556.

(5-Fluoro-2-hydroxyphenyl)(6-phenylpyridin-3-yl)methanone (3g). The title compound (**3g**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 161-163 °C. Yield: 68% (119 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.61 (1H, s), 8.99 (1H, s), 8.09-8.07 (3H, m), 7.90 (1H, d, J = 3.9 Hz), 7.53-7.47 (3H, m), 7.31-7.27 (2H, m), 7.07 (1H, dd, J = 9.0, 4.2 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.1, 160.5, 159.3, 154.7 (d, J = 237.9 Hz), 149.9, 137.8, 137.7, 131.2, 130.3, 129.0, 127.4, 124.5 (d, J = 24.1 Hz), 120.0 (d, J = 8.1 Hz), 120.0, 118.5, 117.6 (d, J = 24.1 Hz); IR (ATR) 3079, 1635, 1582, 1477, 1419, 1329, 1280, 1219, 1131, 1019, 970, 774 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{18}H_{12}FNO_2$: 293.0852. Found: 293.0850.

(5-Chloro-2-hydroxy-4-methylphenyl)(6-phenylpyridin-3-yl)methanone (3h). The title compound (**3h**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 136-138 °C. Yield: 70% (135 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.78 (1H, s), 8.97 (1H, d, J = 1.8 Hz), 8.08 (3H, dd, J = 8.4, 1.8 Hz), 7.90 (1H, d, J = 8.4 Hz), 7.57 (1H, s), 7.53-7.47 (3H, m), 6.98 (1H, s), 2.40 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) δ 197.6, 161.6, 160.3, 149.8, 146.4, 137.7, 132.2, 131.4, 130.2, 129.0, 127.4, 124.5, 120.7, 120.1, 117.9, 20.9; IR (ATR) 2920, 1629, 1587, 1470, 1382, 1290, 1221, 1134, 1019, 859, 780 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{19}H_{14}ClNO_2$: 323.0713. Found: 323.0710.

(*E*)-3-(3-Oxo-3-phenylprop-1-en-1-yl)-4*H*-chromen-4-one (4a). The title compound (**4a**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 134-136 °C. Yield: 15% (25 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.66 (1H, d, J = 15.0 Hz), 8.30 (1H, dd, J = 7.8, 1.2 Hz), 8.18 (1H, s), 8.09-8.08 (2H, m), 7.71-7.68 (1H, m), 7.57-7.55 (1H, m), 7.49-7.47 (3H, m), 7.47-7.44 (2H, m); ^{13}C NMR (150 MHz, $CDCl_3$) δ 190.6 176.3, 158.9, 155.4, 137.9, 135.3, 134.1, 132.9, 128.7, 128.6, 126.3, 125.9, 125.8, 124.3, 119.6, 118.2; IR (ATR) 3080, 3026, 2817, 1664, 1651, 1570, 1496, 1450, 1309, 1287, 1215, 1177, 1078, 989 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{18}H_{12}O_3$: 276.0786. Found: 276.0783.

(2-Hydroxyphenyl)(6-(*m*-tolyl)pyridin-3-yl)methanone (5a). The title compound (**5a**) was prepared according to the general procedure. The product was obtained as a brown solid, mp 66-67 °C. Yield: 77% (133 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.88 (1H, s), 8.97 (1H, s), 8.07 (1H, dd, J = 9.0, 2.4 Hz), 7.91 (1H, s), 7.86 (1H, d, J = 8.4 Hz), 7.84 (1H, d, J = 7.8 Hz), 7.61 (1H, d, J = 7.8 Hz), 7.53 (1H, t, J = 7.8 Hz), 7.39 (1H, t, J = 7.8 Hz), 7.28 (1H, d, J = 7.2 Hz), 7.08 (1H, d, J = 8.4 Hz), 6.91 (1H, t, J = 7.8 Hz), 2.44 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) δ 199.0, 163.2, 160.3, 149.9, 138.7, 137.9, 137.7, 136.8, 132.9, 131.6, 130.9, 128.8, 127.9, 124.4, 119.9, 119.1, 118.9, 118.6, 21.5; IR (ATR) 2920, 1617, 1583, 1477, 1440, 1379, 1333, 1304, 1240, 1141, 1030, 761 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{19}H_{15}NO_2$: 289.1103. Found: 289.1100.

(2-Hydroxyphenyl)(6-(*p*-tolyl)pyridin-3-yl)methanone (5b). The title compound (**5b**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 116-117 °C. Yield: 78% (135 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.89 (1H, s), 8.95 (1H, d, J = 2.4 Hz), 8.04 (1H, dd, J = 7.8, 1.8 Hz), 7.97 (2H, d, J = 7.8 Hz), 7.83 (1H, d, J = 8.4 Hz), 7.61 (1H, dd, J = 8.4, 1.8 Hz), 7.52 (1H, td, J = 8.4, 1.2 Hz), 7.30 (2H, d, J = 7.8 Hz), 7.08 (1H, t, J = 8.4 Hz), 6.90 (1H, td, J = 7.2, 0.6 Hz), 2.41 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.9, 163.1, 160.0, 149.9, 140.4, 137.6, 136.7, 135.1, 132.9, 131.4, 129.7, 119.9, 119.5, 119.1, 118.9, 118.6, 21.3; IR (ATR) 2935, 1619, 1587, 1467, 1445, 1389, 1343, 1309, 1247, 1137, 1032, 765 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{19}H_{15}NO_2$: 289.1103. Found: 289.1101.

(2-Hydroxyphenyl)(6-(4-propylphenyl)pyridin-3-yl)methanone (5c). The title compound (**5c**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 85-87 °C. Yield: 72% (136 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.91 (1H, s), 8.95 (1H, d, J = 8.4, 1.8 Hz), 8.05 (1H, dd, J = 8.4, 1.8 Hz), 7.99 (2H, d, J = 7.8 Hz), 7.84 (1H, d, J = 8.4 Hz), 7.62 (1H, dd, J = 7.8, 1.8 Hz), 7.52 (1H, td, J = 7.2, 1.8 Hz), 7.31 (2H, d, J = 8.4 Hz), 7.08 (1H, d, J = 7.2 Hz), 6.90 (1H, td, J = 8.4, 1.2 Hz), 2.64 (2H, t, J = 7.8 Hz), 1.60 (2H, q, J = 7.8 Hz), 0.95 (3H, t, J = 7.0 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.9, 163.1, 160.1, 149.9, 145.1, 137.7, 136.7, 135.3, 132.9, 131.4, 129.1, 127.2, 119.5, 119.1, 118.9, 118.6, 37.8, 24.3, 13.7; IR (ATR) 2957, 2927, 2867, 1623, 1581, 1478, 1440, 1382, 1329, 1299, 1245, 1219, 1143, 1112, 934, 800, 753 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{19}NO_2$: 317.1416. Found: 317.1413.

(2-Hydroxyphenyl)(6-(4-pentylphenyl)pyridin-3-yl) methanone (5d). The title compound (**5d**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 85-88 °C. Yield: 70% (144 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.92 (1H, s), 8.95 (1H, d, J = 4.4 Hz), 8.04 (1H, dd, J = 8.4, 2.4 Hz), 7.99 (2H, d, J = 7.8 Hz), 7.84 (1H, d, J = 8.4 Hz), 7.61 (1H, dd, J = 7.8, 1.8 Hz), 7.52 (1H, td, J = 8.4, 1.8 Hz), 7.31 (2H, d, J = 7.8 Hz), 7.08 (1H, d, J = 7.2 Hz), 6.90 (1H, td, J = 8.4, 1.8 Hz), 2.66 (2H, t, J = 7.8 Hz), 1.64 (2H, quint, J = 7.8 Hz), 1.35-1.31 (4H, m), 0.88 (3H, t, J = 7.2 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.9, 163.1, 160.1, 149.9, 145.4, 137.6, 136.7, 135.3, 132.9, 131.3, 129.0, 127.2, 119.5, 119.1, 118.9, 118.6, 35.7, 31.4, 30.9, 22.5, 13.9; IR (ATR) 2924, 2855, 1623, 1582, 1476, 1440, 1383, 1329, 1299, 1245, 1144, 935, 753 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{23}H_{23}NO_2$: 345.1729. Found: 345.1728.

(6-(4-Bromophenyl)pyridin-3-yl)(2-hydroxyphenyl)methanone (5e). The title compound (**5e**) was prepared according to the general procedure. The product was obtained as a brown solid, mp 150-152 °C. Yield: 73% (154 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.85 (1H, s), 8.96 (1H, d, J = 1.8 Hz), 8.08 (1H, dd, J = 7.8, 1.8 Hz), 7.95 (2H, d, J = 8.4 Hz), 7.85 (1H, d, J = 7.8 Hz), 7.63 (2H, d, J = 8.4 Hz), 7.59 (1H, dd, J = 8.4, 1.8 Hz), 7.54 (1H, td, J = 8.4, 1.8 Hz), 7.09 (1H, d, J = 8.4 Hz), 6.91 (1H, t, J = 7.8 Hz) ;^{13}C NMR (150 MHz, $CDCl_3$) δ 198.9, 163.2, 158.8, 150.0, 137.8, 136.9, 136.8, 132.9, 132.2, 132.1, 128.8, 124.8, 119.6, 119.1, 119.0, 118.7; IR (ATR) 3065, 1620, 1582, 1465, 1385, 1325, 1280, 1150, 1181, 1021, 834, 754 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{18}H_{12}BrNO_2$: 353.0051. Found: 353.0049.

(2-Hydroxyphenyl)(6-(4-(trifluoromethyl)phenyl)pyridin-3-yl)methanone (5f). The title compound (**5f**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 155-157 °C. Yield: 69% (142 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.85 (1H, s), 9.00 (1H, d, J = 1.2 Hz), 8.19 (2H, d, J = 8.4 Hz), 8.11 (1H, dd, J = 7.8, 1.2 Hz), 7.91 (1H, d, J = 8.4 Hz), 7.67 (2H, d, J = 7.8 Hz), 7.59 (1H, d, J = 8.4 Hz), 7.54 (1H, t, J = 7.2 Hz), 7.09 (1H, d, J = 9.0 Hz), 6.91 (1H, t, J = 7.2 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.8, 163.3, 158.4, 150.1, 141.3, 137.8, 137.1, 132.9, 132.6, 131.7 (q, J = 32.1 Hz), 127.6, 125.9 (q, J = 4.5 Hz), 123.1, 120.2, 119,1, 118.9, 118.8; IR (ATR) 3005, 1620, 1579, 1483, 1437, 1380, 1312, 1239, 1160, 1106, 1064, 1013, 798, 759 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{19}H_{12}F_3NO_2$: 343.0820. Found: 343.0819.

(6-(3,5-Bis(trifluoromethyl)phenyl)pyridin-3-yl)(2-hydroxyphenyl)methanone (5g). The title compound (**5g**) was

prepared according to the general procedure. The product was obtained as a yellow solid, mp 120-122 ºC. Yield: 68% (199 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.82 (1H, s), 9.02 (1H, d, *J* = 1.2 Hz), 8.56 (2H, s), 8.15 (1H, dd, *J* = 8.4, 6.6 Hz), 7.97 (2H, d, *J* = 7.8 Hz), 7.57-7.55 (2H, m), 7.11 (1H, d, *J* = 9.0 Hz), 6.92 (1H, t, *J* = 7.8 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.6, 163.4, 156.5, 150.2, 140.1, 138.1, 137.2, 133.2, 132.8, 132.4 (q, *J* = 33.4 Hz), 127.3 (q, *J* = 3.4 Hz), 123.4 (q, *J* = 3.45 Hz), 123.2 (q, *J* = 271.35 Hz), 120.1, 119.2, 118.9, 118.8; IR (ATR) 2925, 1580, 1485, 1383, 1337, 1275, 1163, 1125, 1071, 835, 790 cm^{-1}; HRMS m/z (M^+) calcd for $C_{20}H_{11}F_6NO_2$: 411.0694. Found: 411.0694.

(6-(4-Ethynylphenyl)pyridin-3-yl)(2-hydroxyphenyl) methanone (5h). The title compound (**5h**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 130-133ºC. Yield: 71% (127 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.85 (1H, s), 8.97 (1H, d, *J* = 1.8 Hz), 8.08 (1H, dd, *J* = 8.4, 2.4 Hz), 8.05 (2H, d, *J* = 9.0 Hz), 7.87 (1H, d, J = 8.4 Hz), 7.62 (2H, d, *J* = 7.8 Hz), 7.59 (1H, dd, *J* = 7.8, 1.2 Hz), 7.55 (1H, td, *J* = 8.4, 1.2 Hz), 7.09 (1H, d, *J* = 7.8 Hz), 6.91 (1H, td, *J* = 7.2, 0.6 Hz), 3.1 (1H, s); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 198.9, 163.2, 158.9, 149.9, 138.1, 137.8, 136.9, 132.9, 132.7, 132.1, 127.2, 123.9, 119.9, 119.1, 119.0, 118.7, 83.2, 79.1; IR (ATR) 3224, 2921, 1619, 1580, 1474, 1442, 1378, 1335, 1300, 1239, 1147, 1030, 832, 758 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{20}H_{13}NO_2$: 299.0946. Found: 299.0946.

(2-Hydroxyphenyl)(6-(naphthalen-1-yl)pyridin-3-yl)me thanone (5i). The title compound (**5i**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 117-119 ºC. Yield: 70% (136 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.92 (1H, s), 9.08 (1H, d, *J* = 1.8 Hz), 8.15-8.13 (2H, m), 7.95 (1H, d, *J* = 8.4 Hz), 7.93-7.92 (1H, m), 7.75 (1H, d, *J* = 7.8 Hz), 7.70 (1H, dd, *J* = 8.4, 1.8 Hz), 7.67 (1H, dd, *J* = 6.6, 1.2 Hz), 7.59-7.50 (4H, m), 7.12 (1H, d, J = 8.4 Hz), 6.95 (1H, td, J = 8.4, 1.2 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 199.2, 163.3, 162.3, 149.7, 137.3, 137.2, 136.9, 133.9, 133.1, 131.8, 130.8, 129.8, 128.5, 127.9, 126.9, 126.1, 125.3, 125.2, 124.6, 119.1, 119.0, 118.7; IR (ATR) 3046, 2921, 2934, 1584, 1483, 1332, 1304, 1239, 1146, 1025, 930, 758 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{22}H_{15}NO_2$: 325.1103. Found: 325.1101.

(2-Hydroxy-5-methylphenyl)(6-(4-methoxy-3-methylph enyl)pyridin-3-yl)methanone (6a). The title compound (**6a**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 130-132 ºC. Yield: 76% (151 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.71 (1H, s), 8.96 (1H, d, *J* = 2.4 Hz), 8.05 (1H, dd, *J* = 7.8, 2.4 Hz), 7.55 (1H, d, *J* = 7.8 Hz), 7.46-7.44 (1H, m), 7.40 (1H, s), 7.35 (1H, dd, *J* = 8.4, 2.4 Hz), 6.99 (1H, d, *J* = 7.8 Hz), 6.85-6.84 (2H, m), 3.84 (3H, s), 2.45 (3H, s), 2.27 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 199.1, 162.6, 161.2, 160.2, 149.4, 137.9, 137.8, 136.9, 132.6, 131.8, 131.4, 131.1, 128.2, 123.6, 118.7, 118.4, 116.5, 111.5, 55.3, 20.9, 20.5; IR (ATR) 3023, 1619, 1583, 1477, 1445, 1385, 1304, 1242, 1209, 1140, 929, 745, 693 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{19}NO_3$: 333.1365. Found: 333.1362.

(6-(3,5-Bis(trifluoromethyl)phenyl)pyridin-3-yl)(2-hydr oxy-5-methylphenyl)methanone (6b). The title compound (**6b**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 140-142 ºC. Yield: 68% (173 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.63 (1H, s), 9.00 (1H, d, *J* = 1.8 Hz), 8.56 (2H, s), 8.15 (1H, dd, *J* = 8.4, 1.8 Hz), 7.98 (2H, dd, *J* = 11.4, 3.6 Hz), 7.37 (1H, dd, *J* = 9.0, 1.2 Hz), 7.32 (1H, s), 7.00 (1H, d, *J* = 4.5 Hz), 2.26 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 198.5, 161.3, 156.4, 150.1, 140.0, 138.3, 138.1, 133.3, 132.4 (q, *J* = 33.4 Hz), 132.3, 128.4, 127.3 (q, *J* = 3.4 Hz), 123.3 (q, *J* = 3.4 Hz), 123.2 (q, *J* = 271.3 Hz), 120.2, 118.6, 20.5; IR (ATR) 2920, 1589, 1482, 1386, 1339, 1277, 1164, 1127, 1070, 830, 791 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{13}F_6NO_2$: 425.0850. Found: 425.0854.

(5-Ethyl-2-hydroxyphenyl)(6-(*p*-tolyl)pyridin-3-yl)meth anone (6c). The title compound (**6c**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 118-119 ºC. Yield: 75% (142 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.74 (1H, s), 8.96 (1H, d, *J* = 1.8 Hz), 8.06 (1H, dd, *J* = 8.4, 1.8 Hz), 7.98 (2H, d, *J* = 7.8 Hz), 7.85 (1H, d, *J* = 7.8 Hz), 7.41 (1H, d, *J* = 1.8 Hz), 7.38 (1H, dd, *J* = 8.4, 1.8 Hz), 7.31 (2H, d, *J* = 8.4 Hz), 7.01 (1H, d, *J* = 8.4 Hz), 2.56. (2H, q, *J* = 7.8 Hz), 2.41 (3H, s), 1.17 (3H, t, *J* = 7.2 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 198.9, 161.3, 160.0, 149.9, 140.3, 137.7, 136.7, 135.2, 134.7, 131.6, 131.5, 129.7, 127.2, 119.6, 118.8, 118.5, 27.9, 21.3, 15.8; IR (ATR) 2967, 2937, 2860, 1623, 1580, 1472, 1450, 1381, 1339, 1290, 1246, 1218, 1142, 1117, 807, 755 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{19}NO_2$: 317.1416. Found: 317.1414.

(5-Bromo-2-hydroxyphenyl)(6-(4-propylphenyl)pyridin -3-yl)methanone (6d). The title compound (**6d**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 110-111 ºC. Yield: 74% (175 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.79 (1H, s), 8.95 (1H, s), 8.05 (1H, dd, *J* = 8.4, 2.4 Hz), 8.00 (2H, d, *J* = 8.4 Hz), 7.86 (1H, d, *J* = 8.4 Hz), 7.72 (1H, d, *J* = 2.4 Hz), 7.59 (1H, dd, *J* = 9.0, 2.4 Hz), 7.31 (2H, d, *J* = 8.4 Hz), 6.99 (1H, d, *J* = 8.4 Hz), 2.65 (2H, t, *J* = 8.4 Hz), 1.68 (2H, q, *J* = 7.2 Hz), 0.95 (3H, t, *J* = 7.8 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 198.0, 162.1, 160.6, 150.0, 145.3, 139.4, 137.6, 135.3, 134.7, 130.7, 129.2, 127.3, 120.7, 120.3, 119.7, 110.6, 37.8, 24.3, 13.8; IR (ATR) 2955, 1617, 1584, 1461, 1371, 1329, 1288, 1210, 1153, 1104, 941, 821, 786 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{21}H_{18}BrNO_2$: 395.0521. Found: 395.0523.

(5-Bromo-2-hydroxyphenyl)(6-(4-bromophenyl)pyridin -3-yl)methanone (6e). The title compound (**6e**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 134-136 ºC. Yield: 75% (193 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.79 (1H, s), 8.95 (1H, d, *J* = 1.8 Hz), 8.06 (1H, dd, *J* = 7.8, 1.8 Hz), 7.96 (2H, d, *J* = 8.4 Hz), 7.86 (1H, d, *J* = 8.4 Hz), 7.69 (1H, d, *J* = 2.4 Hz), 7.63 (2H, d, *J* = 9.0 Hz), 7.61 (1H, dd, *J* = 9.0, 2.4 Hz), 7.00 (1H, d, *J* = 9.0 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 197.9, 162.1, 159.3, 150.0, 139.6, 137.8, 136.7, 134.7, 132.2, 131.4, 128.8, 124.9, 120.8, 120.3, 119.8 110.7; IR (ATR) 3084, 1616, 1579, 1457, 1376, 1323, 1285, 1216, 1149, 1074, 1005, 825, 784 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{18}H_{11}Br_2NO_2$: 430.9157. Found: 430.9160.

(5-Chloro-2-hydroxyphenyl)(6-(*p*-tolyl)pyridin-3-yl)met hanone (6f). The title compound (**6f**) was prepared according to the general procedure. The product was obtained as a brown solid, mp 145-147 ºC. Yield: 74% (143 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.77 (1H, s), 8.95 (1H, s), 8.05 (1H, dd, *J* = 8.4, 1.2 Hz), 7.98 (2H, d, *J* = 7.8 Hz), 7.86 (1H, d, *J* = 9.0 Hz), 7.58 (1H, d, *J* = 2.4 Hz), 7.46 (1H, dd, *J* = 9.0, 2.4 Hz), 7.31 (2H, d, *J* = 7.8 Hz), 7.04 (1H, d, *J* = 9.0 Hz), 2.41 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 198.1, 161.6, 160.5, 149.9, 140.6, 137.6, 136.6, 135.0, 131.7, 130.7, 129.7, 127.2, 123.8, 120.3, 119.7, 119.6, 21.4; IR (ATR) 3291, 3073, 1624, 1586, 1465, 1380, 1323, 1284, 1216, 1168, 1095, 946, 786 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{19}H_{14}ClNO_2$: 323.0713. Found: 323.0712.

(5-Chloro-2-hydroxyphenyl)(6-(3-ethynylphenyl)pyridi n-3-yl)methanone (6g). The title compound (**6g**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 137-139 ºC. Yield: 69% (137 mg). ^{1}H NMR (600 MHz, $CDCl_3$) *δ* 11.74 (1H, s), 8.97 (1H, d, *J* = 1.8 Hz), 8.21 (1H, s), 8.08 (2H, dd, *J* = 8.4, 2.4 Hz), 7.89 (1H, d, *J* = 7.8 Hz), 7.59 (1H, d, *J* = 8.4 Hz), 7.56 (1H, d, *J* = 3.0 Hz), 7.49-7.46 (2H, m), 7.05 (1H, d, *J* = 9.0 Hz), 3.12 (1H, s); ^{13}C NMR (150 MHz, $CDCl_3$) *δ* 198.0, 161.7, 159.4, 149.9, 138.1, 137.7, 136.8, 133.6, 131.7, 131.5, 131.0, 129.1, 127.7, 123.8, 123.0, 120.4, 120.0, 119.6, 83.1, 77.9; IR (ATR) 2912, 1575, 1463, 1396, 1325, 1285, 1214, 1149, 822, 783 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{20}H_{12}ClNO_2$: 333.0557. Found: 333.0555.

(5-Fluoro-2-hydroxyphenyl)(6-(4-pentylphenyl) pyridin -3-yl)methanone (6h). The title compound (**6h**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 121-123 °C. Yield: 72% (156 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.61 (1H, s), 8.96 (1H, d, *J* = 1.8 Hz), 8.05 (1H, dd, *J* = 8.4, 2.4 Hz), 7.99 (2H, d, *J* = 8.4 Hz), 7.85 (1H, d, *J* = 8.4 Hz), 7.32-7.29 (3H, m), 7.28-7.24 (1H, m), 7.05 (1H, dd, *J* = 9.6, 4.8 Hz), 2.66 (2H, t, *J* = 7.8 Hz), 1.65 (2H, quint, *J* = 7.8 Hz), 1.36-1.31 (4H, m), 0.88 (3H, t, *J* = 7.2 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.1, 160.6, 159.3, 154.2 (d, *J* = 239.1 Hz), 149.9, 145.6, 137.5, 135.3, 130.8, 129.1, 127.2, 124.3 (d, *J* = 24.1 Hz), 120.0 (d, *J* = 6.9 Hz), 119.5, 118.5 (d, *J* = 6.9 Hz), 17.6 (d, *J* = 24.1 Hz), 35.7, 31.4, 30.9, 22.5, 13.9; IR (ATR) 2924, 2856, 1633, 1581, 1475, 1418, 1384, 1331, 1277, 1220, 1129, 1016, 1065, 786 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{23}H_{22}FNO_2$: 363.1635. Found: 363.1633.

(5-Fluoro-2-hydroxyphenyl)(6-(4-(trifluoromethyl)phen yl)pyridin-3-yl)methanone (6i). The title compound (**6i**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 133-135 °C. Yield: 75% (162 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.56 (1H, s), 9.01 (1H, d, *J* = 1.8 Hz), 8.20 (2H, d, *J* = 7.8 Hz), 8.11 (1H, dd, *J* = 7.8, 1.8 Hz), 7.93 (1H, d, *J* = 8.4 Hz), 7.77 (2H, d, *J* = 8.4 Hz), 7.31-7.26 (2H, m), 7.08 (1H, dd, *J* = 9.6, 4.8 Hz); ^{13}C NMR (150 MHz, $CDCl_3$) δ 197.9 (d, *J* = 2.4 Hz) 159.5, 158.8, 155.5, 153.9, 149.9, 141.1, 137.8, 132.0, 131.7 (d, *J* = 30.6 Hz), 127.7, 125.9 (q, *J* = 3.4 Hz), 124.7 (q, *J* = 22.9 Hz), 123.1, 120.2 (d, *J* = 6.9 Hz), 118.4 (d, *J* = 5.7 Hz), 117.5 (d, *J* = 22.9 Hz); IR (ATR) 2922, 2854, 1578, 1582, 1476, 1411, 1319, 1220, 1167, 1109, 1065, 1015, 786 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{19}H_{11}F_4NO_2$: 361.0726. Found: 361.0723.

(3,5-Dichloro-2-hydroxyphenyl)(6-(*p*-tolyl)pyridin-3-yl) methanone (6j). The title compound (**6j**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 141-143 °C. Yield: 74% (158 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 12.21 (1H, s), 8.96 (1H, s), 8.08 (1H, d, *J* = 7.8 Hz), 7.99 (2H, d, *J* = 7.8 Hz), 7.89 (1H, d, *J* = 7.8 Hz), 7.63 (1H, s), 7.53 (1H, d, *J* = 7.5 Hz), 7.32 (2H, d, *J* = 7.3 Hz), 2.42 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) δ197.7, 160.8, 157.4, 149.9, 140.9, 137.9, 136.2, 134.7, 130.4, 130.3, 129.8, 127.3, 124.4, 123.7, 120.3, 119.8, 21.4; IR (ATR) 3287, 3173, 1620, 1576, 1475, 1382, 1327, 1280, 1217, 1169, 1099, 947, 789 cm^{-1}; HRMS m/z (M+) calcd for $C_{19}H_{13}Cl_2NO_2$: 357.0323. Found: 357.0323.

(3,5-Dichloro-2-hydroxyphenyl)(6-(4-ethynylphenyl)py ridin-3-yl)methanone (6k). The title compound (**6k**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 157-159 °C. Yield: 71% (156 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 12.19 (1H, s), 8.98 (1H, d, *J* = 1.8 Hz), 8.09 (1H, dd, *J* = 8.4, 2.4 Hz), 8.06 (2H, d, *J* = 8.4 Hz), 7.91 (1H, d, *J* = 8.4 Hz), 7.63 (3H, dd, *J* = 8.4, 2.4 Hz), 7.51 (1H, d, *J* = 2.4 Hz), 3.20 (1H, s); ^{13}C NMR (150 MHz, $CDCl_3$) δ 197.7, 159.8, 157.5, 150.0, 137.9, 137.8, 136.3, 132.8, 130.9, 130.3, 127.3, 124.4, 124.2, 123.7, 120.2, 120.1, 83.1, 79.3; IR (ATR) 2910, 1573, 1460, 1386, 1320, 1275, 1210, 1139, 820, 786 cm^{-1}; HRMS m/z (M+) calcd for $C_{20}H_{11}Cl_2NO_2$: 367.0167. Found: 367.0167.

(5-Chloro-2-hydroxy-4-methylphenyl)(6-(4 (trifluorome thyl)phenyl)pyridin-3-yl)methanone (6l). The title compound (**6l**) was prepared according to the general procedure. The product was obtained as a yellow solid, mp 151-153 °C. Yield: 70% (164 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 11.73 (1H, s), 8.99 (1H, s), 8.20 (2H, d, *J* = 8.4 Hz), 8.10 (1H, dd, *J* = 8.4, 2.4 Hz), 7.93 (1H, d, *J* = 8.4 Hz), 7.77 (2H, d, *J* = 8.4 Hz), 7.54 (1H, s), 6.99 (1H, s), 2.40 (3H, s); ^{13}C NMR (150 MHz, $CDCl_3$) δ 197.5, 161.7, 158.6, 150.9, 149.9, 146.6, 141.2, 137.8, 132.2, 131.8 (q, *J* = 32.1 Hz), 127.7, 125.9 (q, *J* = 3.4 Hz), 124.8 (q, *J* = 270.1 Hz), 124.6, 120.8, 120.4, 117.9, 20.9; IR (ATR) 3009, 1627, 1571, 1480, 1432, 1370, 1301, 1233, 1161, 1100, 1034, 1003, 782, 751 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{20}H_{13}ClF_3NO_2$: 391.0587. Found: 391.0587.

(*E*)-3-(3-Oxohept-1-en-1-yl)-4*H*-chromen-4-one (8). The title compound (**8**) was prepared according to the general procedure. The product was obtained as a white solid, mp 114-120°C.Yield: 46% (70 mg). ^{1}H NMR (600 MHz, $CDCl_3$) δ 8.23 (1H, dd, *J* = 6.6, 1.8 Hz), 8.12 (1H, s), 7.67 (1H, td, *J* = 7.2, 1.2 Hz), 7.56 (1H, d, *J* = 15.6 Hz), 7.45 (1H, d, *J* = 8.4 Hz), 7.42 (1H, t, *J* = 7.8 Hz), 7.28 (1H, d, *J* = 16.2 Hz), 2.60 (2H, t, *J* = 7.2 Hz), 1.62 (2H, quint., *J* = 7.2 Hz), 1.33 (2H, sex., *J* = 7.8 Hz), 0.90 (3H, d, *J* = 7.2 Hz) ; ^{13}C NMR (150 MHz, $CDCl_3$) δ 201.1, 176.1, 157.7, 155.5, 134.0, 132.7, 128.9, 126.3, 125.9, 124.1, 119.4, 118.1, 41.9, 26.3, 22.4, 13.9; IR (ATR) 3050, 3027, 2813, 1660, 1645, 1575, 1490, 1435, 1319, 1288, 1213, 1165, 1071, 989, 780 cm^{-1}; HRMS *m/z* (M^+) calcd for $C_{16}H_{16}O_3$: 256.1099. Found: 256.1096.

Spectrophotometric Measurements:

A UV-Vis spectrophotometer (Optizen UV-3200) was used to evaluate the absorbance spectra of the synthesized compounds at 298K (room temperature). Commercially available **OBZ** (Aldrich, analytical standard, ≥ 98% pure) and synthesized compounds **3a**, **3b**, **3f**, **3h**, **5a**, **5e**, **5f**, **6b**, **6g**, and **6j** were all prepared at a concentration of 50 µM in ethanol solvent (Sigma-Aldrich, ≥99.8% pure). For solvent background, all data were corrected by setting the solvent as blank through the instrument's calibration. As the absorption spectra for UV protecting materials (UVA/UVB) is in the range of 290-400nm, the samples absorption spectra were taken in the range of 200-550 nm. With ethanol as a blank, samples were all prepared in 1 cm quartz cell. The data were obtained in every 5 nm at each point, charted by the data analysis The UVA/UVB-ratio is an arithmetic term of the amount of UVA-coverage with that of UVB-coverage. Another parameter used is critical wavelength λ_c, it is defined as the 90% of the area of absorbance spectrum under the approximate lower wavelength limit of terrestrial sunlight (290 nm) to 400 nm.

Preparation of agar plate for antibacterial studies:

The preparation of agar plate has been included in the supporting information. For the preparation of agar plate, water was added to the nutrient agar mixture powder DifcoTM and the final volume was made up to 500ml in a flask. The flask was covered with a piece of aluminum foil and the contents inside was heated to boil for a 1 min with constant stirring. The contents were then transferred to a 1 L pyrex jar and autoclaved for 20 minutes followed by cooling to ~55 °C and pouring into a thin layer (5mm) of ~8mL into each plate. The plate was swirled in a circular motion in order to distribute the agar on bottom surface uniformly and cooled for about 20-25 min until it settled as a stable gel. The plate was then flipped upside down so as to avoid condensation on the agar medium.

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FULL PAPER

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